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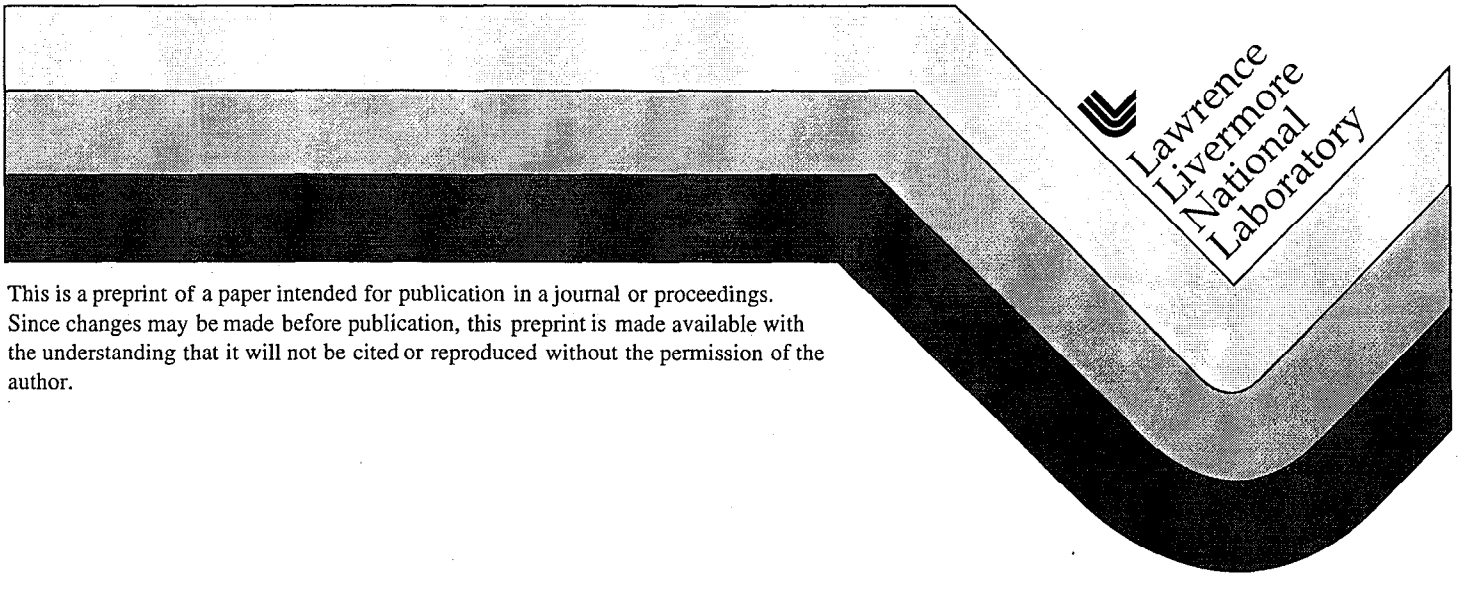

THE PRODUCTION AND DISSOLUTION OF NUCLEAR EXPLOSIVE MELT GLASSES AT UNDERGROUND TEST SITES IN THE PACIFIC REGION*

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Abstract

From 1975 to 1996 the French detonated 140 underground nuclear explosions beneath the atolls of Mururoa and Fangataufa in the South Pacific; from 1965 to 1971 the United States detonated three high yield nuclear tests beneath Amchitka Island in the Aleutian chain. Approximately 800 metric tons of basalt is melted per kiloton of nuclear yield; almost 10^7 metric tons of basalt were melted in these tests. Long-lived and toxic radionuclides are partitioned into the melt glass at the time of explosion and are released by dissolution with seawater under saturated conditions. A glass dissolution model predicts that nuclear melt glasses at these sites will dissolve in 10^6 to 10^7 years.

1. INTRODUCTION

Due to the relative isolation of volcanic islands in the Pacific region, the French Commissariat à l'Energie Atomique and United States Atomic Energy Commission conducted a series of underground nuclear tests on Pacific islands during the period 1965 to 1996. The United States conducted three approximate megaton yield nuclear explosions underground at Amchitka Island in the Aleutian island-arc chain and the French conducted 140 lower yield underground nuclear explosions beneath the atolls of Mururoa and Fangataufa in French Polynesia. All of the tests were conducted in basaltic rocks at depths of burial beneath sea-level. While the detonation of nuclear explosives underground allows for the containment of nearly all prompt radiation residual from the explosion, the subsequent long-term dissolution of radioactive melt glass poses a potential threat to the marine environment. Recently there has been international concern about the potential for radionuclide migration at these sites. Accurate radiological assessments require an understanding of the thermal and mechanical effects of an underground nuclear explosion on submarine basalts, the production and post-shot distribution of radionuclides, the physical and chemical characteristics of the residual matrices, and their long-term performance under saturated conditions.

2. PHENOMENOLOGY

Phenomenology refers to the effects of a nuclear explosion on the surrounding geologic environment. Early-time phenomenology refers to physical and chemical processes that occur immediately after a nuclear detonation during the time the explosion cavity reaches it maximum dimension. Late-time phenomenology refers to thermally-induced process affecting the standing cavity which leads its eventual collapse. Tompson et al. [1] provide a comprehensive summary of phenomenology as it relates to the evolution of a nuclear explosion cavity and collapse chimney.

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2.1. Early-time phenomenology

When an underground nuclear explosion is detonated, sufficient energy is released to instantaneously vaporize rock adjacent to the firing point. Initial temperatures are several million °K and pressures are close to 1 Mbar. A compressive, outwardly propagating shock wave is generated within a millisecond which crushes, melts, and vaporizes rock creating an expanding open volume or cavity. As the geologic media surrounding the explosion point is melted, pore water is also vaporized creating steam in the still growing cavity; CO_2 may also be produced from the fusion of carbonate materials. Depending on the explosive yield the cavity reaches its maximum size within 80 to 500 milliseconds. A compressive hoop stress is created by the rebound of the adjacent media; this residual stress will tend to close radial fractures that were created during the initial expansion of the cavity. The final size of the standing cavity is a function of the explosive yield, the density of the surrounding rock and the overburden stress. Based on underground experience, the French provide an empirical measure for maximum cavity dimension at Mururoa and Fangataufa:

$$R_c = 100\ (Y/h)^{1/3} \qquad (1)$$

where

R_c is the cavity radius in meters,
Y is the total yield in kilotons,
h is depth of device burial in meters.

2.2. Late-time phenomenology

After the shock waves and elastic stress waves have dissipated, less volatile vapors begin to condense on the walls of the standing cavity in a molten state. The melt follows down the walls of the cavity and drips from the roof forming a puddle on the cavity bottom. For basalts with an average density of 2.5 g/cm^3 the French report that approximately 800 metric tons of rock are melted per kiloton of nuclear yield [2]. Water vapor, noncondensible gases (such as CO_2 and H_2) and other noncondensed radionuclides (such as tritiated water vapor, ^{85}Kr, and ^{137}Xe) are still present as gases in the cavity. Within minutes to hours after the detonation, thermal energy in the gas is conductively transferred through the walls of the cavity to the surrounding geologic media creating high thermal gradients. These thermal stresses induce ablation and spall of wall rock which falls into the cavity quenching the melt.

Within minutes to hours debris continues to fill the cavity. Condensation of gas causes a reduction in cavity pressure until it reaches a few bars. When the pressure in the cavity drops below surrounding ambient lithostatic pressure, the cavity collapses and propagates upwards forming a collapse chimney. If the explosive yield is low, the rock strength is high, or the test is deeply buried, the collapse chimney will not reach the ground surface.

3. RADIONUCLIDE PRODUCTION AND DISTRIBUTION

For a generic thermonuclear device, the radioactivity residual from an underground nuclear is derived from five components:

(1) Residue of nuclear fuel materials (e.g.,^{3}H, ^{6}LiD, ^{235}U, ^{238}U, ^{239}Pu, ^{240}U),
(2) Reaction products of fuel materials (e.g., ^{3}H, ^{237}Np, ^{239}Pu, ^{240}Pu, ^{241}Am),

(3) Fission products (e.g., ^{85}Kr, ^{90}Sr, ^{99}Tc, ^{125}Sb, ^{129}I, ^{137}Cs, ^{155}Eu),
(4) Activation products on non-fuel device materials (e.g., ^{14}C, ^{60}Co, ^{55}Fe), and
(5) Activation products of backfill materials and adjacent geology (e.g., ^{36}Cl, ^{41}Ca, ^{152}Eu).

The production of selected radionuclides is presented in Table I.

TABLE I. PRODUCTION AND PARTITIONING (IN %) BETWEEN GLASS, DEBRIS RUBBLE, GAS AND WATER OF SELECTED RADIONUCLIDES PRODUCED DURING AN UNDERGROUND NUCLEAR EXPLOSION [3]

Nuclide	Decay	Half-Life (a)	Production	Glass	Rubble	Gas	Water
^{3}H	β^-	12.33	fuel residue and fuel product	0	0	2	98
^{14}C	β^-	5730	(n,p) in device	0	10	80	10
^{85}Kr	β^-	10.77	fission product	0	10	80	10
^{90}Sr	β^-	28.78	fission product	40	60	0	0
^{99}Tc	β^-	211100	fission product	80	20	0	0
^{129}I	β^-	15700000	fission product	50	40	0	10
^{137}Cs	β^-	30.07	fission product	40	60	0	0
^{152}Eu	EC	13.54	(n, γ) in environment	95	5	0	0
^{155}Eu	β^-	4.76	fission product	95	5	0	0
^{237}Np	α	2144000	fuel residue and fuel product	95	5	0	0
^{239}Pu	α	24110	fuel residue and fuel product	98	2	0	0
^{241}Am	α	432	fuel residue and fuel product	98	2	0	0

Refractory radionuclides with higher boiling points and lower vapor pressures (i.e., Zr, Tc, Ce, Eu, U, Pu, Am, Cm) are largely volumetrically incorporated in the puddle glass. Radionuclides with lower boiling points and higher vapor pressures (H, Sr, Ru, Sb, Cs, Ba) are heterogeneously distributed throughout the cavity-chimney system as a result of volatilization, displacement, and condensation accompanying cavity collapse. Volatile radionuclides will largely condense on fracture and mineral surfaces in addition to partitioning in the glass. The fractionation of radionuclides has also been documented on field-scale. Post-shot drilling into a nuclear test cavity-chimney have returned samples which provide a nearly continuous profile of radioactivity ascending from the cavity upwards through the collapse chimney [4]. Refractory radionuclides (e.g., ^{152}Eu, ^{154}Eu) are concentrated in the puddle glass while ^{3}H and volatile fission products (e.g., ^{137}Cs) are found in the higher chimney away from the working point. Table I shows the general partitioning of radionuclides (in %) between melt glass, crystalline rubble and breccia, water, and gas phases for selected radionuclides residual from underground nuclear tests.

4. GLASSES PRODUCED DURING AN UNDERGROUND NUCLEAR EXPLOSION

Nuclear explosive melt glass is produced from the condensation of vaporized rock, the shock melting of native rock, and the melting of neighboring rock in contact with the melt and vapor [5].

These glasses are significant for two reasons:

(1) Glasses volumetrically incorporate radionuclides which are subsequently released by matrix dissolution under saturated or partially saturated conditions.
(2) The nuclear explosive melt glasses partition actinide species (i.e., ^{235}U, ^{238}U, ^{237}Np, ^{239}Pu, ^{240}U) which have higher toxicities (organ dose conversion factors: $\sim 10^{-6}$ to $\sim 10^{-5}$ Sv/Bq) and half-lives sufficiently long to dominate the residual source term inventory after ~ 200 years (i.e., $t_{1/2}$ of ^{239}Pu is 2.41 x 10^4 years, $t_{1/2}$ of ^{237}Np is 2.14 x 10^6 years).

The glass produced during an underground nuclear explosion is extremely heterogeneous and includes:

(1) Dark, dense highly radioactive glass which is representative of the puddle accumulation of melt.
(2) Light, variably vesicular glass with less radioactivity which is representative of melted host rock.
(3) Globs, belbs, and coatings of dark glass which formed from the rapid condensation of vaporized silicates on fracture surfaces [6].

Autoradiographic and petrographic analyses of three samples of glass from a single test confirm the presence of a dark vesicular glass with a high alpha track density as well as a less vesicular lighter colored mixed glass with a lower fission track density [7]. The major and trace element composition of the glass mimics that of the host rock from which it both vaporized and melted [6]. In general, the melt glass is heterogeneous containing both mineral and lithic inclusions. Quenching is non-uniform. For these reasons, melt glass is both variably charged with fine vesicles and fractured.

4.1. Glass produced beneath Mururoa, Fangataufa, and Amchitka

The French have detonated 140 underground nuclear explosions (exclusive of safety trials) beneath the atolls of Mururoa and Fangataufa in French Polynesia during the period 1975 to 1996 [2]. The total yield of all the French underground tests is ~3,200 kilotons [3]. On the Aleutian Islands in the Bering Sea the United States detonated three high yield nuclear explosions: LONGSHOT on 29 October 1965 with a total yield of ~ 80 kilotons, MILROW on 2 October 1969 with a yield of ~ 1000 kilotons, and CANNIKIN on 6 November 1971 with a total yield < 5000 kilotons [8]. The firing medium for all these explosions was basalt produced by volcanism associated with the formation of ocean islands (hot spots and arcs). The SiO_2 concentration of basalt from the CANNIKIN emplacement hole is 44.2 weight % and the SiO_2 content of Mururoa basalts is 44.8 weight % SiO_2. The bulk density of the South Pacific and Amchitka basalts is 2.5 g/cm^3.

As noted above, for rocks of this density the French report 800 metric tons of basalt is melted per kiloton of total nuclear yield. Assuming a total yield for the French tests on Mururoa and Fangataufa of 3200 kilotons and a maximum (upper limit) total yield of United States nuclear tests on Amchitka of 6080 kilotons, 2.56 x 10^6 metric tons of melt glass is estimated to be beneath Mururoa and Fangataufa and 4.86 x 10^6 metric tons is estimated to be beneath Amchitka.

5. GLASS DISSOLUTION

Glasses are thermodynamically unstable matrices which transform with time to stable crystalline phases. Water acts as a flux to allow the transformation; the rate of the transformation provides an upper limit to release rates of radioactive elements contained in the glass. Glass reactions in aqueous environments begin with water diffusion into the glass and alkali ion exchange. Hydration and ion exchange on the glass surface result in an inner diffusion layer several microns thick where alkalis and water form distinct concentration gradients and an outer, hydrated gel layer where Si network dissolution occurs [9]. The layer migrates into the glass releasing elements stoichiometrically. Most of the elements are quickly incorporated into alteration phases which include clays, zeolites, oxides, and hydroxides.

In general the dissolution of glass can be described by a rate equation which relates the dissolution of glass per unit time to a rate constant for dissolution, the reactive surface area of the glass and an affinity term that represents the thermodynamic driving force for dissolution of the solid phase. A simplified rate equation may be written:

$$dn/dt = S\ k\ (1-Q/K) \qquad (2)$$

where

n is grams of glass released,
t is the unit time,
S is the reactive surface area,
k is the forward rate constant for dissolution,
Q is the concentration product for the dissolving solid in solution,
K is the solubility product for the dissolving solid.

As species originally present in the glass builds up in solution, the dissolution rate decreases. Previous studies of silicate glasses have shown that higher concentrations of dissolved silica slows the reaction rate. Because of lack of more detailed information, a glass dissolution model has been developed that calculates only the release of silica from the matrix. A complete description of the model is provided by Tompson et al. [1]. The rate of dissolution of nuclear explosive melt glass may be calculated as a function of the temperature of reaction, the mass of glass dissolved, the SiO_2 content of the glass, the SiO_2 content of the reacting solution, the pH of the solution, the density of the glass, and the surface area of the glass. Using a temperature of 30°C, 45 weight % SiO_2 in the basalt, 10 mg/L Si in the reacting fluid, and a pH of 8.0, the model calculates a dissolution rate of 1.56×10^{-3} grams of glass/m^2/day. This rate compares favorably with dissolution rates (7.26×10^{-4} grams/m^2/day to 7.26×10^{-3} grams/m^2/day) obtained from experimental studies of the reaction of Mururoa basalts and free silicon seawater calculated at 25°C [10].

Using 8000 metric tons of glass produced for a 10 kiloton explosion, a melt glass porosity of 30%, and an empirically derived glass cracking factor of 25 which relates the total surface areas to the geometric surface area, the model calculates a melt glass surface area of 0.07 cm^2/g. The resulting final reaction rate for the glass is 9.3×10^{1} g/day. For a 100 kiloton yield explosion, the final reaction rate will increase to 4.3×10^{2} g/day. For a 1000 kiloton yield explosion, the rate rises to 2.0×10^{3} g/day. Using the 10 kiloton rate (9.3×10^{1} g/day) for the glass beneath Mururoa and Fangataufa, all the glass will dissolve in 7.5×10^{7} years. Using the 1000 kiloton rate (2.0×10^{3} g/day) for Amchitka, all the glass will dissolve in 6.7×10^{6} years.

The hydraulic conductivity of the Mururoa and Fangataufa massive basalts ranges between 10^{-8} and 10^{-7} m/sec facilitating glass-water reactions [11].

6. CONCLUSIONS

The above calculations apply only to the dissolution of melt glass. It should be emphasized that transport of radionuclides through the near-field involves a variety of processes in addition to dissolution which may include ion-exchange, precipitation, complexation, sorption, and formation of, or sorption to, colloids [12]. As well, radionuclide concentrations will be dispersed and diluted by seawater. Over an interval of 10^6 to 10^7 years, ^{239}Pu will decay in excess of 300 half-lives. The release rates calculated by the model are extremely sensitive to the temperature, pH, porosity, and specific surface area assumed for dissolution of the glass. In particular measurement of specific surface area is difficult on matrices of these kind, and this variable significantly affects the final reaction rate [13].

Estimates of environmental contamination due to the dissolution of nuclear explosive melt glass produced beneath French and U.S. underground nuclear weapons test sites in the Aleutians and South Pacific are influenced by these factors:

(1) After several hundred years, relatively toxic and long-lived radionuclides (i.e., U, Pu, Am, Np, Cm) dominate the residual radionuclide source term.
(2) These radionuclides are volumetrically partitioned in nuclear explosive melt glasses.
(3) All the glasses produced are both saturated and soluble.

Predictions based on current models suggest dissolution rates are slow enough that constituent radionuclides will be released from the melt glass for more than 10^6 to 10^7 years.

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